

A simple cross-correlation technique between infrared and hard x-ray pulses

Bertold Krässig,^{a)} R. W. Dunford, E. P. Kanter, E. C. Landahl,^{b)} S. H. Southworth, and L. Young

Argonne National Laboratory, Argonne, Illinois 60439, USA

(Received 18 February 2009; accepted 1 April 2009; published online 30 April 2009)

We report a gas phase technique to establish the temporal overlap of ultrafast infrared laser and hard x-ray pulses. We use tunnel ionization of a closed shell atom in the strong field at the focus of an infrared laser beam to open a distinct x-ray absorption resonance channel with a clear fluorescence signature. The technique has an intrinsic response of a few femtoseconds and is nondestructive to the two beams. It provides a step-functionlike cross-correlation result. The details of the transient provide a diagnostic of the temporal overlap of the two pulses. © 2009 American Institute of Physics. [DOI: 10.1063/1.3125256]

Monitoring atomic motions on their intrinsic time and length scales (angstroms and femtoseconds) represents a great opportunity for x-ray studies of matter. An important class of studies involves understanding the response of atoms, molecules and materials to photoexcitation. Here the key technical challenge is ensuring the overlap between a hard x-ray pulse derived from a free-electron laser (FEL) or synchrotron radiation source and an optical pulse derived from a separate laser. Cross correlation of two optical pulses is routine using nonlinear crystals, but cross correlation between an optical and hard x-ray pulse (where hard x rays refer to photon energies >4 keV) has so far only been demonstrated using x-ray diffraction based methods.^{1,2} In these methods the ultimate response time is governed by the dynamical response of the crystalline material. The absorption of an ultrafast optical pulse generates a coherent strain wave propagating at the speed of sound (~ 4000 m/s) that modifies the x-ray diffraction intensity, yielding a Bragg peak change in a few picoseconds. A cross correlator with an initial time response approaching tens of femtoseconds and applicability to hard x rays has been demonstrated at the Hamburg FEL (FLASH) (Ref. 3) with the aid of electro-optic sampling (EOS).⁴ The response is governed by the ultrafast electron rearrangement following x-ray absorption within a 10 nm surface layer of GaAs that alters optical reflectivity. Here we demonstrate a gas phase technique that has the potential for an intrinsic response time of a few femtoseconds. We use optical-field ionization of krypton gas followed by x-ray resonant absorption to provide a simple step function cross correlation. Our method is closely related to that of Ref. 5, who use the transient state of Kr^+ ions during optical-field ionization as an ultrafast soft x-ray absorption sampling gate.

An alternative, nonresonant, gas phase cross-correlation method has been demonstrated for optical and EUV pulses with photon energies <100 eV.^{6–8} In two-color photoionization, the photoelectrons ejected by the EUV pulse exchange energy with the optical laser “dressing” field, producing sidebands on the main photoelectron peak. These sidebands ap-

pear only in the presence of both the optical and EUV fields, and thus the cross-correlation signal yields a convolution of the optical and EUV pulse shapes and their relative jitter. Impressive results have been obtained at FLASH in Xe at an EUV photon energy of 90 eV using a magnetic bottle spectrometer with 4π collection efficiency.⁹ The implementation of the sideband technique for the hard x-ray regime is made difficult by the combination of low photoionization cross sections, gas density restrictions for electron spectroscopy, and lifetime broadening of the x-ray photoelectron line.

Our infrared/x-ray cross correlation is a pump-probe technique utilizing two intrinsically ultrafast atomic physics processes: Strong optical field ($\sim 10^{14}$ W/cm²) ionization of closed-shell atoms as a pump and resonant x-ray fluorescence in the resulting ions as a probe. The use of initially closed shell atoms assures good contrast because the resonant photoexcitation channel is only present in the ionized species. In this work we are interested in cross-correlation measurements with hard x rays, focusing on core excitations and subsequent fluorescence emission, using krypton as an example. X-ray fluorescence has the practical advantages of being penetrating and unaffected by external fields. The same principles apply to other closed-shell systems and resonant excitations from other shells, as long as the resonant states decay promptly with a readily observable signature. For neutral Kr the x-ray absorption spectrum is an essentially featureless step, blending the pre-edge Rydberg series of $1s$ excitations and the actual $1s$ ionization edge (Fig. 1, top). In contrast, the additional $1s \rightarrow 4p$ resonance in Kr^+ is well separated and virtually background free (Fig. 1, bottom). The signature of the promotion of a $1s$ electron is the prompt emission of a fluorescence photon. The time scales determining the response time of this technique are very short: the optical-field ionization occurs during a few optical cycles (2.67 fs period for 800 nm), depending on the peak electric field strength. For example, in the central core of the focus of 2 mJ (30 μJ) laser pulses with 50 fs (5 fs) full width at half maximum (FWHM) duration, focused to 100 μm (30 μm) FWHM diameter, ionization of krypton atoms is saturated within six (1.5) optical cycles,¹⁰ generating an almost pure sample of Kr^+ . The lifetime of the $1s$ -hole state is 0.24 fs and the main decay mechanism is fluorescence emission. While the time scales for the creation of the Kr^+ ion cloud and the

^{a)}Electronic mail: kraessig@anl.gov.

^{b)}Present address: Department of Physics, De Paul University, Chicago, IL 60614.

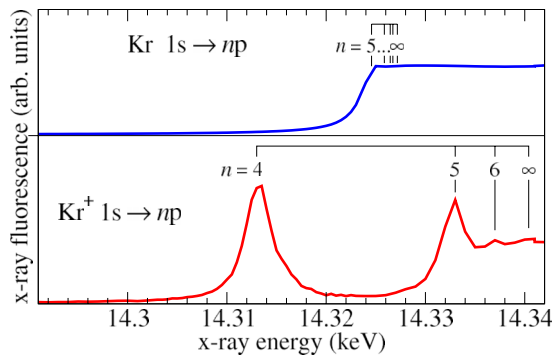


FIG. 1. (Color online) Fluorescence yield signal from x-ray absorption by neutral Kr atoms (upper trace) and Kr^+ ions (lower trace) as a function of x-ray energy. The energies corresponding to resonant excitations are indicated.

appearance of the Kr^+ signature in x-ray absorption are very short, the ion cloud and the existence of the resonant absorption persist for a significantly longer time after the ionizing laser pulse and weaken with a 10 ns time constant because of the relatively slow Coulomb expansion of the Kr^+/e^- ensemble. Neutral atoms in the interaction region are replenished at the speed of sound, $0.34 \mu\text{m}/\text{ns}$. The atomic physics aspects of this process and the associated plasma dynamics have been the subject of several recent publications^{11–15} and are well understood. In terms of the temporal resolution, the practical limitations of the technique come from the jitter in the synchronization between laser and x-ray pulses and the accuracy of their relative delay setting.

The experimental setup and procedure used in this work are described in detail in Ref. 14. Briefly, a monochromatized x-ray beam from Argonne's Advanced Photon Source (APS) is focused to about $10 \times 10 \mu\text{m}^2$ and intersected with an intense Ti:Sapphire (2 W, 50 fs, 1 kHz) laser beam focused to $100 \times 100 \mu\text{m}^2$ at a crossing angle of 25 mrad inside a vacuum chamber containing krypton gas (Fig. 2). The laser oscillator is phase-locked to the APS driving radio frequency and the relative delay is controlled by a variable delay line. A pair of x-ray fluorescence detectors is mounted at right angles to the x-ray beam and 1 mm slits limit the detector viewing length to the section of complete beam overlap. Locating the smaller x-ray focus fully inside the laser focus and restricting the detector viewing range minimizes the spatial averaging of laser intensity for the probed volume. The strongest signal is obtained for linear laser polarization in the same plane as the x-ray polarization. The signal is half that strength if the polarization directions are at right angles due to the orbital alignment in the tunneling process.^{13–15} The alignment decreases with time and this decrease is the more rapid the higher the gas density.¹¹ In contrast, the overall signal decrease stemming from the Cou-

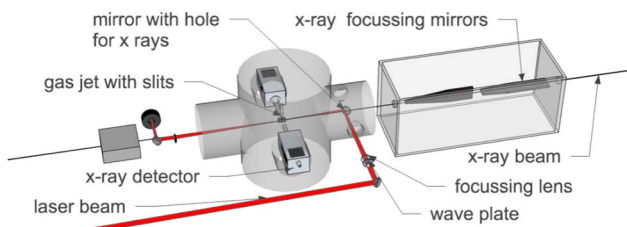


FIG. 2. (Color online) Experimental setup.

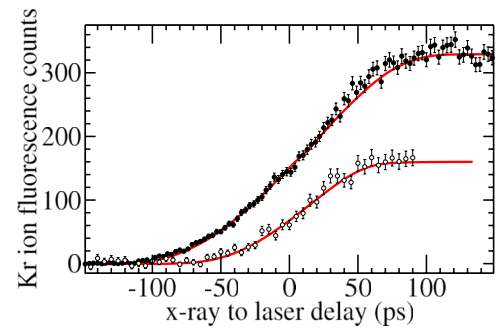


FIG. 3. (Color online) Cross-correlation data for hybrid fill (solid circles, 10 s/ch) and 24-bunch mode (open circles, 20 s/ch). A background from neutrals (~ 10) was subtracted. The solid curves are the cumulative integral of the respective x-ray pulse profiles from streak camera measurements.

lomb expansion of the ion cloud is independent of the gas density.¹²

In our cross-correlation measurement the x-ray energy is tuned to the $\text{Kr}^+ 1s \rightarrow 4p$ resonance energy. The 1 kHz laser pulses are overlapped with a particular single bunch in the APS fill pattern once for every 271 revolutions. We record fluorescence counts originating in the region of spatial overlap from this particular bunch, separately with and without laser, while stepping the relative delay between the x-ray and laser pulses. The observed transient in the fluorescence signal versus relative delay is mathematically the convolution of the stepwise ion buildup in the oscillating laser field with the x-ray temporal pulse shape and with the jitter in the laser-x-ray synchronization. In the present case the temporal width of the ion buildup (~ 15 fs) and the jitter (~ 5 ps) rms are small compared to the width of the x-ray pulse (~ 100 ps), so the observed delay dependence can be regarded as the cumulative integral of the x-ray pulse shape.

The steplike cross correlation from an irreversible process such as the present case has the practical advantage that one can perform and optimize the spatial overlap with the laser timing set coarsely to within a few nanoseconds earlier than the x-rays using a photodiode, and then finding the temporal overlap by decreasing the relative delay until the fluorescence signal is reduced to 50%. Cross-correlation techniques based on reversible processes, e.g., the generation of sidebands,⁹ require simultaneous spatial and temporal overlap to yield a signal.

Figure 3 shows results for resonant x-ray fluorescence emitted as a function of the x-ray-to-laser delay. The data were obtained during “24-bunch” and “hybrid fill” operations at the APS. The laser polarization was at 45° to the x-ray polarization in order to eliminate the pressure dependent signal decrease due to dealignment. At large negative delays the x-ray pulse precedes the laser pulse and no ion fluorescence is observed. Superimposed on the data are numerically integrated x-ray pulse profiles measured with the APS diagnostic streak camera at a different time.¹⁶ In hybrid mode our data indicate near the top of the figure a somewhat steeper rise in the transient than seen in the streak camera results. We show in Fig. 4 the results of x-ray bunch length determinations from various cross-correlation measurements at the APS performed over a period of four years.

In this letter we present a resonant gas phase cross-correlation technique between ultrafast infrared and hard x-ray pulses with an inherent response time of a few optical

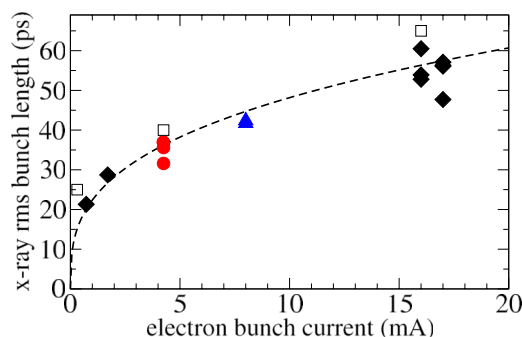


FIG. 4. (Color online) Root mean square (rms) x-ray bunch lengths extracted from cross-correlation measurements as a function of the electron bunch current. Diamonds, hybrid fill, 16/17 mA injected; the two points below 2 mA were taken after an extended period without top-up injections. Triangles, hybrid fill 8 mA. Circles, 24-bunch, 4 mA. Open squares, APS nominal x-ray bunch lengths. The dashed curve represents a cube root functional behavior.

cycles. Because of the low number density of target atoms the technique is nondestructive for the two beams and applications as an in-line diagnostic could be conceived—with the obvious limitation to specific resonant x-ray energies. With improvements in the relative jitter between the two beams to levels as reported in,⁹ this method would be well suited to characterize proposed picosecond x-ray pulse implementations.¹⁷ At future x-ray FELs the x-ray fluxes will suffice to perform single-shot measurements with our method at a given delay. Schemes have been proposed to reduce the x-ray pulse length at FELs to below 1 fs.¹⁸ The synchronization of such short x-ray pulses to an external laser system in pump/probe applications may not be achievable at this level and one will have to measure the x-ray arrival time on a shot-to-shot basis analogously to EOS for the electron bunches.⁴ For that case, and for scattering/diffraction/imaging applications that typically don't require continuous x-ray tunability, one could imagine tailoring the intensity of the ionizing infrared laser in the present method such that the buildup of the ion sample is stretched over a few 100 s of femtoseconds, effectively providing a streaking mechanism which would map the observed fluorescence yield to the delay between the subfemtosecond x-ray and ionizing laser pulses.

This work was supported by the Chemical Sciences, Geosciences, and Biosciences Division, (and also in the case of the Advanced Photon Source) the Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy under Contract No. DE-AC02-06CH11357.

- ¹A. M. Lindenberg, I. Kang, S. L. Johnson, T. Missalla, P. A. Heimann, Z. Chang, J. Larsson, P. H. Bucksbaum, H. C. Kapteyn, H. A. Padmore, R. W. Lee, J. S. Wark, and R. W. Falcone, *Phys. Rev. Lett.* **84**, 111 (2000).
- ²D. A. Reis, M. F. DeCamp, P. H. Bucksbaum, R. Clarke, E. Dufresne, M. Hertlein, R. Merlin, R. Falcone, H. Kapteyn, M. M. Murnane, J. Larsson, T. Missalla, and J. S. Wark, *Phys. Rev. Lett.* **86**, 3072 (2001).
- ³C. Gahl, A. Azima, M. Beye, M. Deppe, K. Döbrich, U. Hasslinger, F. Hennies, A. Melnikov, M. Nagasono, A. Pietzsch, M. Wolf, W. Wurth, and A. Föhlisch, *Nat. Photonics* **2**, 165 (2008).
- ⁴A. L. Cavalieri, D. M. Fritz, S. H. Lee, P. H. Bucksbaum, D. A. Reis, J. Rudati, D. M. Mills, P. H. Fuoss, G. B. Stephenson, C. C. Kao, D. P. Siddons, D. P. Lowney, A. G. MacPhee, D. Weinstein, R. W. Falcone, R. Pahl, J. Als-Nielsen, C. Blome, S. Düsterer, R. Ischebeck, H. Schlarb, H. Schulte-Schrepping, T. Tschentscher, J. Schneider, O. Hignette, F. Sette, K. Sokolowski-Tinten, H. N. Chapman, R. W. Lee, T. N. Hansen, O. Synnørgren, J. Larsson, S. Techert, J. Sheppard, J. S. Wark, M. Bergh, C. Caleman, G. Hultdt, D. van der Spoel, N. Timneanu, J. Hajdu, R. A. Akre, E. Bong, P. Emma, P. Krejcik, J. Arthur, S. Brennan, K. J. Gaffney, A. M. Lindenberg, K. Luening, and J. B. Hastings, *Phys. Rev. Lett.* **94**, 114801 (2005).
- ⁵K. Oguri, T. Nishikawa, T. Ozaki, and H. Nakano, *Opt. Lett.* **29**, 1279 (2004).
- ⁶T. E. Glover, R. W. Schoenlein, A. H. Chin, and C. V. Shank, *Phys. Rev. Lett.* **76**, 2468 (1996).
- ⁷M. Meyer, D. Cubaynes, P. O'Keeffe, H. Luna, P. Yeates, E. T. Kennedy, J. T. Costello, P. Orr, R. Taïeb, A. Maquet, S. Düsterer, P. Radcliffe, H. Redlin, A. Azima, E. Plönjes, and J. Feldhaus, *Phys. Rev. A* **74**, 011401 (2006).
- ⁸S. Cunovic, N. Müller, R. Kalms, M. Krikunova, M. Wieland, M. Drescher, T. Maltezopoulos, U. Frühling, H. Redlin, E. Plönjes-Palm, and J. Feldhaus, *Appl. Phys. Lett.* **90**, 121112 (2007).
- ⁹P. Radcliffe, S. Düsterer, A. Azima, H. Redlin, J. Feldhaus, J. Dardis, K. Kavanagh, H. Luna, J. P. Gutierrez, P. Yeates, E. T. Kennedy, J. T. Costello, A. Delserieys, C. L. S. Lewis, R. Taïeb, A. Maquet, D. Cubaynes, and M. Meyer, *Appl. Phys. Lett.* **90**, 131108 (2007).
- ¹⁰R. Santra, personal communication (3 February 2009).
- ¹¹C. Höhr, E. R. Peterson, N. Rohringer, J. Rudati, D. A. Arms, E. M. Dufresne, R. W. Dunford, D. L. Ederer, E. P. Kanter, B. Krässig, E. C. Landahl, R. Santra, S. H. Southworth, and L. Young, *Phys. Rev. A* **75**, 011403 (2007).
- ¹²E. P. Kanter, R. Santra, C. Höhr, E. R. Peterson, J. Rudati, D. A. Arms, E. M. Dufresne, R. W. Dunford, D. L. Ederer, B. Krässig, E. C. Landahl, S. H. Southworth, and L. Young, *J. Appl. Phys.* **104**, 073307 (2008).
- ¹³R. Santra, R. W. Dunford, and L. Young, *Phys. Rev. A* **74**, 043403 (2006).
- ¹⁴S. H. Southworth, D. A. Arms, E. M. Dufresne, R. W. Dunford, D. L. Ederer, C. Höhr, E. P. Kanter, B. Krässig, E. C. Landahl, E. R. Peterson, J. Rudati, R. Santra, D. A. Walko, and L. Young, *Phys. Rev. A* **76**, 043421 (2007).
- ¹⁵L. Young, D. A. Arms, E. M. Dufresne, R. W. Dunford, D. L. Ederer, C. Höhr, E. P. Kanter, B. Krässig, E. C. Landahl, E. R. Peterson, J. Rudati, R. Santra, and S. H. Southworth, *Phys. Rev. Lett.* **97**, 083601 (2006).
- ¹⁶B. Yang and Y.-C. Chae, personal communication (15 October 2008).
- ¹⁷A. Zholents, P. Heimann, M. Zolotarev, and J. Byrd, *Nucl. Instrum. Methods Phys. Res. A* **425**, 385 (1999).
- ¹⁸S. Reiche, P. Musumeci, C. Pellegrini, and J. Rosenzweig, *Nucl. Instrum. Methods Phys. Res. A* **593**, 45 (2008), and references therein.